## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

410-009-0

In Re Application Of:

NORBERT BUSCH ET AL

Serial Number: 015,752

Filed: February 27, 1979

For: AN ETHER OF N-PROPANOL

AMINE

Group Art Unit 122

Examiner: TOVAR

REISSUE APPLICATION FOR UNITED STATES LETTERS
PATENT SUPPLEMENTAL DECLARATION
AND POWER OF ATTORNEY

HONORABLE COMMISSIONER OF PATENTS AND TRADEMARKS Washington, D. C. 20231

Sir:

We, Norbert BUSCH, Jacque SIMOND, Andre MONTEIL, Jacque MOLEYRE and Roland Yves MAUVERNAY, citizens of FRANCE, residing, respectively, at LeBouquet 63410-Loubeyrat; 25 Bld Thermal 63400- Chamalieres; HLM "Le Patural" Bat. H. Alee 167 63360-Gerzat; 21 rue Sarrazin 63200-Mozac and 13 rue E. Gilbert 63200-Riom,

hereby declare

that we verily believe that we are the original, first and joint inventors of the invention described and claimed in Letters Patent Number 3,962,238, issued on June 8, 1976 and in Reissue application Serial Number 015,752, filed February 27, 1979, entitled "An Ether Of N-Propanol Amine" and for which we solicit a patent; that we do not know and do not believe that said invention was ever known or used before our invention or discovery thereof; that we acknowledge our duty to disclose information of which we are aware which is material to the examination of this application;

that, while we believe the process for preparing the compounds of the invention is operative, we believe that the ptent is wholly or partly inoperative by reason of a defective specification.

It has been determined, particularly by mass spectroscopic analysis, that the structure of compound I of the present invention, resulting from the reaction of 1-(3-isobutoxy-2-chloro)propyl pyrrolidine and N-benzylaniline results in a compound of the following structure:

rather than the structure originally disclosed, to wit:

SPEC

while the molecular weight of compounds of both structures, above, (based on the empirical formula for these compounds being  $C_{24}H_{34}N_20$ ) is the same, the mass spectral analysis of the compound, as prepared above, shows a weak molecular ion at m/e of 366.2620 (confirming the molecular formula) but, significantly, also shows fragments at m/e 196 ( $C_{14}H_{14}N$ ) and m/e 170 ( $C_{10}H_{20}N0$ ). These fragments can only arise from the former structure, above, to wit:

The mechanism of reaction of the starting compounds most probably proceeds through an intermediate immonium ion which, theoretically could result in two products as shown below:

immonium ion 
$$CH_2$$
  $CH - OCH_2 - CH$   $CH_3$   $CH_2$   $CH_2$   $CH_2$   $CH_3$   $CH_2$   $CH_2$   $CH_3$   $CH_2$   $CH_2$   $CH_3$   $CH_2$   $CH_3$   $CH_2$   $CH_3$   $CH_2$   $CH_3$   $CH_3$   $CH_3$   $CH_4$   $CH_5$   $CH_5$ 

However, while theproducts resulting from the process of the present invention have the same utility and most physical analytical data remain unchanged, mass spectral analysis shows the correct structure to have resulted via cleavage at (a).

Consequently, new Claims 7 and 8 replace all the granted claims to specifically correct this error and, while the utility and physical constants of the other compounds as originally claimed in U.S. Patent No. 3,962,238 remain the same, their structural formulae are open to question, and the claims have been restricted to one compound only, the structure of which has been confirmed by mass spectral analysis as shown above.

The error or errors which led to a defective specification arose in the following manner:

When we started our research with chloramines of type (I),

$$\begin{array}{c} \text{CH}_{3} \\ \text{CH}_{2} - \text{O} - \text{CH}_{2} - \text{CH} - \text{CH}_{2} - \text{N} \\ \text{C1} \end{array}$$

we considered the possibilities of immonium ion (III) attack.

$$\begin{array}{c}
 & \bigoplus_{\text{CH}_2} \text{CH} - \text{CH}_2 - \text{O} - \text{CH}_2 - \text{CH}_2 - \text{CH}_3 \\
 & \bigoplus_{\text{CH}_3} \text{CH}_3
\end{array}$$
(III)

The first series of compounds derived from chloramines (I) were obtained by the following reaction scheme:

Ar 
$$C = 0 + R - 0 - CH_2 - CH - CH_2 - N$$

NH<sub>3</sub> liq. Cl

Na

which could yield either (IV) or (V).

By dehydration of the products, we were in a position to establish the following structure:

since nuclear magnetic resonance spectra did not show the ethylenic proton which would have been present had the type (V) structures been obtained

This led us to assign the structure (IV) to the above-mentioned amino alcohols.

When type (I) chloramines are allowed to react with sodium diphenyl methane in THF, compounds of type (VIII) are obtained.

These are similar to those resulting from catalytic hydrogenation of type (VI) compounds.

In the cases mentioned above, the reactions led to unique compounds not to mixtures of isomers, although mixtures are obtained when type (I) chloramines are allowed to react with sodium derivatives of benzonitrile or diethylmalonate.

The above-mentioned data and the fact that NMR and IR spectra were in agreement with the structure envisaged for compound I of the present invention led us to consider that we were indeed dealing with the compound of structure:

$$CH_3$$
 $CH - CH_2 - O - CH_2 - CH - CH_2 - N$ 
 $CH_2$ 
 $CH_2$ 

In addition, this presumption was reinforced by several published works describing the reactions of type (I) chloramines:

- a) "Synthesis of new aryloxy derivatives of N-substituted propylamines."
  - V. Danksas, G. Pikunaile Zh. Vses. Khim. Obshestsva 9(3), 352 354, 1964.

$$\bigcirc \longrightarrow 0 - CH_2 - CH - CH_2 - N$$

$$\bigcirc \longrightarrow 0 - CH_2 - CH - CH_2 - N$$

$$\bigcirc \longrightarrow 0 - CH_2 - CH - CH_2 - N$$

$$\bigcirc \longrightarrow 0 - CH_2 - CH - CH_2 - N$$

$$\bigcirc \longrightarrow 0 - CH_2 - CH - CH_2 - N$$

$$\bigcirc \longrightarrow 0 - CH_2 - CH - CH_2 - N$$

$$\bigcirc \longrightarrow 0 - CH_2 - CH - CH_2 - N$$

$$\bigcirc \longrightarrow 0 - CH_2 - CH - CH_2 - N$$

b) S. Mamedov et al Zh. Organ. Khim. <u>2</u>(8), p. 1377-1382, 1966 -C.A. 66 54966

$$C_{6}H_{13} - O - CH_{2} - CH - CH_{2} - N$$
Et

 $C_{1}$ 
 $C_{1}$ 
 $C_{1}$ 
 $C_{1}$ 
 $C_{1}$ 
 $C_{1}$ 
 $C_{1}$ 

The above described error in Letters Patent No. 3,962,238 arose without any fraudulent or deceptive intention on our part; and the above is a true specification of the error.

We hereby appoint Norman F. Oblon, Registration Number 24,618, Stanley P. Fisher, Registration Number 24,344, Marvin J. Spivak, Registration Number 24,913, C. Irvin McClelland, Registration Number 21,124, Gregory J. Maier, Registration Number 25,599, Arthur I. Neustadt, Registration Number 24,854, Robert C. Miller, Registration Number 25,357 and Richard D. Kelly, Registration Number 27,757, our (my) attorneys, with full powers of substitution and revocation, to prosecute this application and to transact all business in the Patent Office connected therewith; and we (I) hereby request that all correspondence regarding this application be sent to the firm of OBLON, FISHER, SPIVAK, McCLELLAND & MAIER, P.C., whose Post Office Address is: Crystal Square Five - Suite 400, 1755 South Jefferson Davis Highway, Arlington, Virginia 22202.

We, the undersigned, declare further that all statements made herein of our own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made

are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: April 28, 1980

Date: April 28, 1980

Signature: Signature: Sacque Simond

Jacque Simond

Date: April 28, 1980

Date: April 28, 1980

Signature: Facque Montell

Date: April 28, 1980

Signature: Facque Moleyre

Date: April 22, 1980

Signature: Facque Moleyre

The assignee of Letters Patent No. 3,962,238, hereby indicates by the signature and seal below, the approval of the filing of this reissue application.

Date: April 18, 1980

Signature: /

63200° ATO W

Title: Chairman of the Board